Atty. Docket #: AG-6564

### IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

INTERNATIONAL APPL, NO.: PCT/EP99/05147:

INTERNATIONAL FILING DATE: -07/20/1999-:

APPLICANT: KLAUS-PETER CRONE ET AL

SERIAL NO:

ART UNIT:

FILED:

-HEREWITH-

**EXAMINER:** 

FOR:

"METHOD OF PRODUCING

SOLAR CELLS"

:

Commissioner of Patents

Box PCT

Washington, D.C. 20231

\*Express Mail\* No.: EE617838642

Date: -JANUARY 29, 2001-

I hereby certify that this paper, along with any other paper or fee referred to in this paper as being transmitted herewith, is being deposited with the United States Postal Service "Express Mail Post Office to Addressee" service under 37 CFR 1.10, postage prepaid, on the date indicated above, addressed to the Asst. Comm. for Patents, Washington, D.C. 20231

-Carrie A. McPherson-(Typed or printed name of mailing paper or fee) (Signature of person mailing paper)

TRANSMITTAL OF APPLICATION PAPERS
TO U.S. DESIGNATED/ELECTED OFFICE (DO/EO/US)
CONCERNING A FILING UNDER 35 U.S.C. §371

(37 CFR 1.494 OR 1.495)

This Transmittal Letter is based upon PTO Form 1390 (as revised in May, 1993).

The above-identified applicant(s) (jointly with their assignee) have filed an International Application under the P.C.T. and hereby submit(s) to the United States Designated/Elected Office (DO/EO/US) the following items and other information.

has NOT expired.

d. [] have not been made and will not be made.

e. [] will be submitted with the appropriate surcharge.

AG-6564

JC02 Rec'd PCT/PTO 2 9 JAN 2001

- 1. Ma This is a FIRST submission of items concerning a filing under 35 U.S.C. §371. 2. [] This is a SECOND or SUBSEQUENT submission of items concerning a filing under 35 U.S.C. §371. 3. X This is an express request to begin national examination procedures (35 U.S.C. §371[f]) at any time rather than delay. 4. X A proper Demand for International Preliminary Examination (IPE) was made to the appropriate Authority (IPEA) within the time period required. 5. X A copy of the International Application as filed (35 U.S.C. §371[c][2]) -a. [X] is transmitted herewith (required when not transmitted by International Bureau) -in English-. b. [] has been transmitted by the International Bureau. See WIPO Publication c. [] is not required, as the application was filed in the United States Receiving Office (RO/US). 6. X A (verified) translation of the International Application into the English language is enclosed. (See above). 7. [] Amendments to the (specification and) claims of the International Application under PCT Article 19 (35 U.S.C. 371[c][3]) a. [] are transmitted herewith (required if not transmitted by the International Bureau). b. [] have been transmitted by the International Bureau. c. [] have not been made; however, the time limit for making such amendments
- 8. [] A translation of the amendments to the claims (and/or the specification) under PCT Article 19 (35 U.S.C. §371[c][3]) is enclosed or will be submitted with the appropriate surcharge.

AG-6564

## JC02 Rec'd PCT/PTO 2 9 JAN 2001

9.	[X] An oath or	r declaration/power	of attorney	of the	inventor(s)	(35	U.S.C.
	§371[c][4]) w	ill follow.					
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[ ] and is attached to the translation of (or a copy of) the International Application.

[] and is attached to the substitute specification.

10. [X] A translation of at least the Annexes to the IPE Report under PCT Article 36 (35 U.S.C. §371[c][5]) is enclosed. No annexes a figure of the dead to IPER, and Not mentioned on IPER

Items 11. to 16. below concern other document(s) or information included:

- 11. [] An Information Disclosure Statement under 37 CFR 1.97 and 1.98 is enclosed.
- 12. [X] An Assignment for recording and a separate cover sheet in compliance with 37 CFR 3.28 and 3.31 will follow.
- 13. [X] A FIRST preliminary amendment is enclosed.

  A SECOND or SUBSEQUENT preliminary amendment is enclosed.
- 14. [] A substitute specification (including claims, abstract, drawing) is enclosed.
- 15. [] A change of power of attorney and/or address letter is enclosed.
- 16. [X] Other items of information:
  - This application is being filed pursuant to 37 CFR 1.494(c) or 1.495(c), and any missing parts will be filed before expiration of-
    - [ 22 months from the priority date under 37 CFR 1.494(c), or
    - [X] 32 months from the priority date under 37 CFR 1.495(c).
  - The undersigned attorney is authorized by the International applicant and by the inventors to enter the National Phase pursuant to 37 CFR 1.494(c) or 1.495(c).

The following additional information relates to the International Application:

# International Application No. PCT/EP99/05147

AG-6564

Receiving Office: EPO M

IPEA (if filing under 37 CFR 1.495): EPO X

Priority Claim(s) (35 USC §§ 119, 365): M

German Appln. 198 34 358.2 filed -July 30, 1998-.

A copy of the International Search Report is

enclosed.

x attached to the copy of the International Application.

A copy of the Receiving Office Request Form is enclosed. X

The fee calculation is set forth on the next page of this Transmittal Letter.

[X] PCT/IB/308 (1) sheet

[X] PTO/SB/05 (1) sheet

### FEE CALCULATION SHEET

A check in payment of the filing fee, calculated as follows, is attached (37 CFR 1.492).

Basic Fee	\$ 860.00
Total Number of claims in excess of (20) times \$18	-0-
Number of independent claims in excess of (3) times \$80	-0-
Fee for multiple dependent claims \$270	-0-

TOTAL FILING FEE...

86:0.00

Kindly send us the official filing receipt.

The Commissioner is hereby authorized to charge <u>any</u> additional fees which may be required or to credit any overpayment to Deposit Account No. 03-2775. This is a "general authorization" under 37 CFR 1.25(b), except that no <u>automatic</u> debit of the issue upon allowance is authorized. An additional copy of this page is attached.

Respectfully submitted,

ASHLEY I. PEZZNER

Reg. No. 35,646

CONNOLLY BOVE LODGE & HUTZ LLP

1220 Market Street

P.O. Box 2207

Wilmington, Delaware 19899

Tel. (302) 658-9141

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(Only for new	nonprovisional applications under 37 C.I	F.R. § 1.53(b)) Exp	oress Mail Label No.	EE61	7838642	
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See MPEP ct	hapter 600 concerning utility patent appli		ADDRESS	TO: Box Pater	nt Application	
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	Statement Regarding Fed sponsore Reference to Microfiche Appendix	MR&D			ying identity of above cop	
	Background of the Invention				PLICATION PARTS	162
	Brief Summary of the Invention					
	Brief Description of the Drawings (if Detailed Description	filed)	37.CF	R.§3.73(b) Stat	over sheet & document(s))	)
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Name	Ashley I. Pezz					
	CONNOLLY BOVE		HUTZ LLP		· · · · · · · · · · · · · · · · · · ·	
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City	Wilmington		DE		10000	
Country	US	State	DE (302) 888 - 69	Zip Code	19899	
	US	Telephone	(302)888-62	2/0   Fax	(302)656-907	2

Signature

Date 01/29/2001

Burden Hour Statement: This form is estimated to take 622 hours to complete. Time will vary depending upon the needs of the individual case. Any comments on the amount of time you are required to complete this form should be sent to the Chief Information Officer, Patent and Trademark Office. Washington, DC 20231. DO NOT SEND FEES OR COMPLETED FORMS TO THIS ADDRESS. SEND TO: Assistant Commissioner for Patents, Box Patent Application, Washington, DC 20231.

Registration No. (Attorney/Agent)

35,646

ASHLEY I. PEZZNER

Name (Print/Type)

METHOD OF **PRODUCING** SOLAR CELLS

Klaus-Peter Crone Günter Löffelmann Karl Modemann Eberhard Koch -and-Wolfgang Sauerteig

INTERNATIONAL APPLICATION IN ENGLISH -with-SEARCH REPORT

PCT/EP99/05147

IFD: 07/20/1999

AG-6564

(7244\*87)

\*Express Mail\* mailing tabel number <u>EE617</u>838642 Date of Deposit -January 29, 2001

I hereby certify that this paper or fee is being deposited with the United States Postal Service "Express Mai! Post Office to Assresse' service under 37CFR 1 10 on the date undicates above and is addressed to Box PCT, Commissioner for Patents, Washington. D.C. 20231
—Carrie A. McPherson—
(Typed or printed name of person mailing

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(Signature of person mailing paper of fee)

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# IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

APPLICANT: KLAUS-PETER CRONE E	TAL.
SERIAL NO. TO BE ASSIGNED	) ART UNIT: TO BE ASSIGNED
FILED: HEREWITH	) EXAMINER: TO BE ASSIGNED
FOR: METHOD OF PRODUCING SOLA	AR CELLS )
Asst. Commissioner for Patents	,
Washington, D.C. 20231	
"Express Mail" No. EE617838642 Date January	19,3001
1 HEREBY CERTIFY THAT THIS PAPER OR FEE IS BEING DEPOSIT	ED WITH THE UNITED STATES POSTAL SERVICE "EXPRESS MAIL POST
OFFICE TO ADDRESSEE" SERVICE UNDER 37 CFR 1.10 ON THI	DATE INDICATED AND IS ADDRESSED TO THE
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### **PRELIMINARY AMENDMENT**

Sir:

Prior to fee calculation and examination please amend the above-identified application as follows.

### In the Claims

Amend claim 1 as follows:

In claim 1, line 8, please delete "in particular 400 to 600°C,".

Please add the following new claim(s).

--7. The method as claimed in claim 1, wherein the coating is carried out at temperature from 400 to 600°C. - -

### **REMARKS**

The applicants respectfully request that the preliminary amendment be entered prior to fee calculation and examination. Support for newly added claim 7 can be found in original claim 1. No fee is due for the additional claim. If there are any additional fees due in connection with the filing of this response, the Commissioner is authorized to charge or credit any overpayment to Deposit Account No. 03-2775.

A prompt and favorable action is solicited.

Respectfully submitted,

CONNOLLY BOVE LODGE & HUTZ LLP

Bv

Ashley I Pezzner Reg. No. 35,646

Tel. (302) 888-6270

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PCT/EP99/05147

### Method of producing solar cells

The invention relates to an economically improved method of producing thin-layer solar cells, e.g. CdTe solar cells (CdTe = cadmium telluride). CdTe is used below merely as an example for all thin-layer solar cells.

CdTe and CdTe/CdS solar cells may be produced by various methods (US-5 304 499), common to all of which is heat treatment at at least 575°C, to achieve adequate efficiency. These temperatures allow the use only of expensive types of glass as supports. The use of glass as a support has the disadvantage that glass panels may be coated with CdTe only in discontinuous manner, irrespective of the coating method selected.

US-5 304 499 describes a method in which coating is carried out at temperatures of only 480 to 520°C, it thus being possible to use cheap types of glass ("window glass").

To this end, it is necessary for the glass firstly to be provided with a transparent, electrically conductive layer, e.g. of doped tin oxide. This is followed by a thin cadmium sulfide layer (CdS), to which the light-sensitive CdTe layer is then applied by sublimation at 480 to 520°C.

The apparatus required for application of the CdTe layer is complex and expensive: support material and CdTe source are held in such a way by opposing graphite blocks, which are heated to the necessary temperature, that the CdTe source is located only 2 to 3 mm from the support surface. Sublimation is then effected in a 0.1 mbar inert gas atmosphere, e.g. a nitrogen, helium, argon or hydrogen atmosphere. Large areas of CdTe-coated material for producing solar cells cannot be produced economically in this way.

H. Nishiwaki et al, Solar Energy Materials and Solar Cells 37 (1995) 295 to 306 use a polyimide film as support because this material has sufficient heat resistance in

comparison to polyethylene terephthalate (PET) and polyethylene naphthalate (PEN), because of its glass transition temperature of more than 500°C. Polyimide has the disadvantage to be insoluble in usual solvents and to be unable to be molten. It is therefore extremely difficult to be processed.

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The known methods do not permit the use of easily produceable films of polymeric organic materials as supports.

The object of the invention was the economic production of a support with a photovoltaically active layer, e.g. a CdTe layer.

A method was surprisingly found, which permits the use of flexible polymeric films for coating with CdTe and annealing, without the polymeric supporting material being damaged by the high temperatures. In this way, a starting material is obtained for high efficiency solar cells.

The invention therefore provides a method of coating organic polymeric supporting materials with CdTe and annealing the CdTe layer of the materials thus coated, characterised in that the supporting material consists of a polymeric material with a glass transition temperature of from 90°C to 200°C and coating of the CdTe layer is carried out at temperatures below the glass transition temperature and annealing at temperatures of at least 250°C, in particular 400 to 600°C, by means of a laser for 0.01 to 1 s with an energy of 2 to 5000 watt per mm<sup>2</sup>.

25 Preferably the supporting material is at least 60  $\mu$ m, in particular 90 to 120  $\mu$ m, thick and the CdTe layer is at most 30  $\mu$ m, in particular 2 to 7  $\mu$ m, thick.

Coating is carried out for example with an aqueous or solvent-containing CdTe suspension.

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The material is then dried. Suitable coating methods are, for example, flooding and, knife coating.

Annealing may be carried out several times; cooling phases are preferably provided between pairs of annealing steps.

Suitable polymers are PET and PEN. Prior to coating, the polymeric supporting material may be provided with a substrate layer, e.g. of indium-tin oxide, which improves the adhesion of the CdTe layer. The substrate layer should be transparent and electrically conductive.

Suitable lasers are, for example, argon lasers and yag lasers with frequency duplication.

Organic polymeric supporting materials are flexible and thus permit continuous coating using a suitable coating method.

It is especially advantageous for the CdTe particles to be particularly fine, in particular in the form of so-called nano-particles, i.e. particles whose average diameter lies in the nanometric range and amounts, for example, to from 3 to 5 nm.

In this case, it is expedient for an agent to be present during production of the nanoparticles which prevents agglomeration of the nanoparticles, e.g. tributyl-phosphane.

The invention also provides a solar cell comprising at least one CdTe layer at most 30 µm thick on a support, characterised in that the support is a polymeric organic material at least 60 µm thick and having a glass transition temperature of at least 90°C.

Owing to its flexibility, the polymeric organic support permits continuous coating by means of a coater, for example a meniscus or curtain coater, as known from the coating of photographic films.

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### Example

A film of PEN 100 μm thick and 100 cm wide is coated continuously with a suspension containing a dispersant and 31 g of cadmium telluride per litre. The coated film is then dried and the layer applied exhibits a dry layer thickness of 5 μm.

The film is annealed as follows:

The-entire surface is irradiated with an Ar ion laser (wavelength 514 nm; power 7 W) with a focal point of 50 μm. The temperature is adjusted at from 400 to 450°C.

After annealing, the film exhibits light-dependent electrical resistance and is thus suitable for the production of a photovoltaic cell.

The supporting material is not damaged by exposure to the laser.

### Claims

1. A method of coating organic polymeric supporting materials with at least one photovoltaically active layer and annealing the materials thus coated, characterised in that the supporting material consists of a polymeric material with a glass transition temperature of from 90°Cto 200°C. Coating is carried out at temperatures below the glass transition temperature and annealing at temperatures of at least 250°, in particular 400 to 600°C, by means of a laser for 0.01 to 1 s with an energy to 2 to 5000 watt per mm<sup>2</sup>.

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- 2. A method according to claim 1, wherein the supporting material is at least 60 µm thick and the photovoltaic layer is at most 30 µm thick.
- 3. A method according to claim 1, wherein the supporting material is of polyethylene terephthalate or polyethylene naphthalate.
  - 4. A solar cell comprising at least one photovoltaically active layer on a support, characterised in that the support is a polymeric organic material having a glass transition temperature of from 90°C to 200°C.

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- 5. A solar cell, according to claim 4, characterised in that the photovoltaically active layer comprises cadmium telluride.
- 6. A solar cell according to claim 4, characterised in that the support consists of polyethylene terephthalate or polyethylene naphthalate.

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Attorney Docket No.

AG 6564 (7244\*87)

As a below named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated below next to may name,

believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if ural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention

**動**titled METHOD FOR PRODUCING SOLAR CELLS

specification of which check one)

☐ is attached hereto.

was filed on 07/20/99 as International Patent Application PCT/EP99/05147 and including all the amendments through the date hereof

Application Serial No. 09/744,722 and

was amended on\_ (if applicable) was amended through

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose to the Office all information known to me to be material to patentability as defined in Title 37, Code of Federal Regulations, §1.56.

I hereby claim foreign priority benefits under 35 U.S.C. §119(a)-(d) or 365(b) of any foreign application(s) for patent or inventor's certificate, or §365(a) of any PCT International application which designated at least one country other than the United States, listed below and have also identified below, by checking the box, any foreign application for patent or inventor's certificate, or PCT International application having a filing date before that of the application on which priority is claimed:

Prior Foreign Applica	tion(s)	<u>Pri</u>	Priority Claimed  10/02/00 ⊠ □  (Day/Month/Year Filed) Yes No			
WO 00/07250 (Number)	World (Country)					
(Number)	(Country)	(Day/Month/Year Filed)	□ □ □ Yes No			
(Number)	(Country)	(Day/Month/Year Filed)	_ □ □ □ Yes No			

I hereby claim the benefit under Title 35, United States Code, § 120 of any United States application(s) listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States application in the manner provided by the first paragraph of Title 35, United States Code, § 112, I acknowledge the duty to disclose to the Office all information known to me to be material to patentability as defined in Title 37, Code of Federal Regulations, § 1.56 which became available between the filing date of the prior application and the national or PCT international filing date of this application

(Application Serial No.)	(Filing Date)	(Status) (patented, pending, abandoned)
(Application Serial No.)	(Filing Date)	(Status) (patented, pending, abandoned

I hereby claim the benefit under 35 U.S.C. §119(e) of any United States provisional application(s) listed below.

(Application No.) (filing date)

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.



Telephone: 703-305-3752

UNITED STATES DEPARTMENT OF COMMERCE
Patent and Trademark Office
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U.S. APPLICATION NO		FIRST NAMED APPLICANT	ATTY DOCKET NO
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CONNOLLY BOVE LODGE & H			INTERNATIONAL APPLICATION NO
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NOTIFICATION OF I	MISSING REQUIREMENTS	UNDER 35 U.S.C. 3	71 IN THE UNITED
1. The following items have been s	TES DESIGNATED/ELECTE ubmitted by the applicant or the	IB to the United State	'US) es Patent and Trademark Office as
☐ a Designated Office	e (37 CFR 1.494),		
an Elected Office ( U.S. Basic National Fee.	37 CFR 1.495):		
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Other: RO 101, IB 306, 308  2. The following items <b>MUST</b> be for	urnished within the period set for	orth below in order to	complete the requirements for
acceptance under 35 U.S.C. 371:			
a. Translation of the applicat appropriate 20 or 30 months	tion into English. Note a proce-	ssing fee will be requi	red if submitted later than the
		sons indicated on th	ne attached Notice of Defective
Translation.			
30 months from the priority	ng the translation of the applica date (37 CFR 1.492(f)).	tion and/or the Annex	es later than the appropriate 20 or
C. Oath or declaration of the	inventors, in compliance with	37 CFR 1.497(a) and (	(b), identifying the application by
	number and international filing declaration does not comply wi		nd (h) for the reasons indicated
on the attached PCT		m 5/ CFR 1.49/(a) ai	id (b) for the reasons indicated
	ne oath or declaration later than	the appropriate 20 or	30 months from the priority date
(37 CFR 1.492(e)). 3. Additional claim fees of \$	as a large entity	emall entity includin	g any required multiple dependent
claim fee, are required. Applicant n	nust submit the additional claim	fees or cancel the add	litional claims for which fees are
due. See attached PTO-875.			
ALL OF THE ITEMS SET FORT	H IN 2(a)-2(d) AND 3 ABOV	E MUST BE SUBMIT	TTED WITHIN ONE MONTH
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CFR 1.136(a).	dended by raing a potition and	· ·	ne under the provisions of 57
4. Translation of the Annexes MUS	T be submitted no later that the	time period set above	or the annexes will be cancelled
Note processing fee will be required			
5. The Article 19 amendments ar	e cancelled since a translation v	vas not provided by the	e appropriate 20 (37 CFR.
494(d)) or 30 (37 CFR 1.495(d)) mo	nins from the priority date.		
Applicant is reminded that any commaddress given in the heading and incl			
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FORM PCT/DO/EO/905 (December	[ 199 <i>1)</i>	ı elepn	one: 703-305-3752

POWER OF ATTORNEY: As a named inventor, I hereby appoint the following attorney(s) and/or agent(s) to prosecute this application and transact all business in the Patent and Trademark Office connected therewith:

In the matter of the above-identified application, please recognize Rudolf E. Hutz, Reg. No. 22,397; John D. Farchild, Reg. No. 19,756; Harold Pezzner, Reg. No. 22,112; Richard M. Beck, Reg. No. 22,580; Paul E. Crawford, Reg. No. 24,397; Thomas M. Meshbesher, Reg. No. 25,083; Patricia Smink Rogowski, Reg. No. 33,791; Robert G. McMorrow, Jr., Reg. No. 30,962; Ashley I. Pezzner, Reg. No. 35,646; William E. McShane, Reg. No. 32,707; Mary W. Bourke, Reg. No. 30,982; Gerard M. O'Rourke, Reg. No. 39,794; Allan N. Kutzenco, Reg. No. 38,945; James M. Olsen, Reg. No. 40,408; Francis DiGiovanni, Reg. No. 37,310; J. Kenneth Joung, Reg. No. 41,881; Frank Z. Yang, Reg. No. 35,417; and Eric J. Evain, Reg. No. 42,517 all of P.O. Box 2207, Wilmington, Delaware 19899-2007 as attorneys with full power of substitution to prosecution this application and conduct all business in the Patent and Trademark Office connected the rewith...

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